Radioprotection 2017, 52(1), 29–36
© EDP Sciences 2017
DOI: 10.1051/radiopro/2016085

Available online at:
www.radioprotection.org

ARTICLE

Assessment of ^{137}Cs contamination of combustion products and air pollution during the forest fires in zones of radioactive contamination

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Received: 23 January 2016 / Accepted: 28 November 2016

Abstract – The results of the investigation of radiation hazard of forest fires in the radioactive contaminated areas in the article are presented. The activity concentration of long-lived radionuclides in forest fuel materials is shown. Airborne concentration of ^{137}Cs produced by the combustion of forest fuel materials with different contamination density can exceed the permissible activity levels of ^{137}Cs in air. During the large wildfires on the territories with high values of radioactive contamination density, the airborne concentration of radionuclides may be three or four times higher than the background values. Seasonal fluctuations of average volume activity were estimated by using the ratio between the values of aerosols activity in the atmosphere on the day of the fire (fire season) and the reference values.

Keywords: forest fires / radioactive aerosols / seasonal fluctuations / activity index

1 Introduction

After the Chernobyl Nuclear Power Plant (ChNPP) accident in 1986, more than 4 million hectares of forestland in Belarus, Ukraine and Russia were contaminated by radionuclides. In Belarus, more than 1.7 million hectares of forested areas having soil contamination density exceeding 37 kBq m^{-2} were contaminated by ^{137}Cs. The southern part of Gomel region was also highly contaminated by long-lived radionuclides such as plutonium (^{238}Pu, ^{239,240}Pu) with the half-life time ranging around one hundred to thousands of years, ^{241}Am and ^{90}Sr with the half-life time 432.8 and 29.1 years, respectively. In 2014, the area of radioactively contaminated forests in Gomel region was about 49% of total forestland area. Pine trees cover more than 60% of forestland area in the Gomel region. According to a lot of research studies (Fawaris and Johanson, 1994; Pietrzak-Flis *et al.*, 1996; Yoshida *et al.*, 2004), the forest litter retains most of ^{137}Cs activity in a forest phytocenosis. The contribution of litter to the total radioactive contamination of phytocenosis can range from 40% to 70% (Teramage *et al.*, 2014).

The threat of forest fires revealed after numerous fires in the Chernobyl exclusion zone (CEZ) in 1992 on the border with Ukrainian part of exclusion zone (Hao *et al.*, 2009). As a result of large forest fire at the territory of Polesie State Radiation-Ecological Reserve (Gomel Region, Belarus) in 1992 the level of ^{137}Cs in smoke aerosols was increased 10 times within the 30 km exclusion zone (Dusha-Gudym, 2005). Several studies have noted that during the forest fire in CEZ in May 1992 volume activity of ^{137}Cs in the air has increased by two orders of magnitude compared with normal values (about 3×10^{-3} Bq m^{-3}) (Lujaniene *et al.*, 1997; Yoschenko *et al.*, 2006). The large amount of wildfires in Belarus also have been registered in 1996, 1999 and 2002. Between 1993 and 2013 over 1147 of forest and grasslands fires occurred in the CEZ (Zibtsev *et al.*, 2015).

During forest fires on contaminated territories, radionuclides deposited in forest fuel materials are released in the air with smoke. Radioactive smoke plumes may have relatively high values of volume activity, which is able to exceed the allowable levels of radioactivity in the air. The emission containing radionuclides deposited on the fine aerosol fractions is particularly dangerous. ^{137}Cs and ^{90}Sr are the main dose-forming isotopes after 30 years after the Chernobyl disaster. These radionuclides are dangerous both for external exposure and for internal exposure pathway (inhalation and ingestion) (Hohl *et al.*, 2012). Inhalation of radionuclides (especially ^{238}Pu, ^{239,240}Pu, ^{241}Am) can generate additional internal doses both for firefighters, close to the ignition source, and for citizens far from the source. In addition, radionuclides released from the fire source may be transferred with the airflows over long distances, which can lead to a secondary radioactive contamination, which may be significant especially when it spreads over territories that do not exhibit high contamination level, *i.e.*, that remain safe of contamination.

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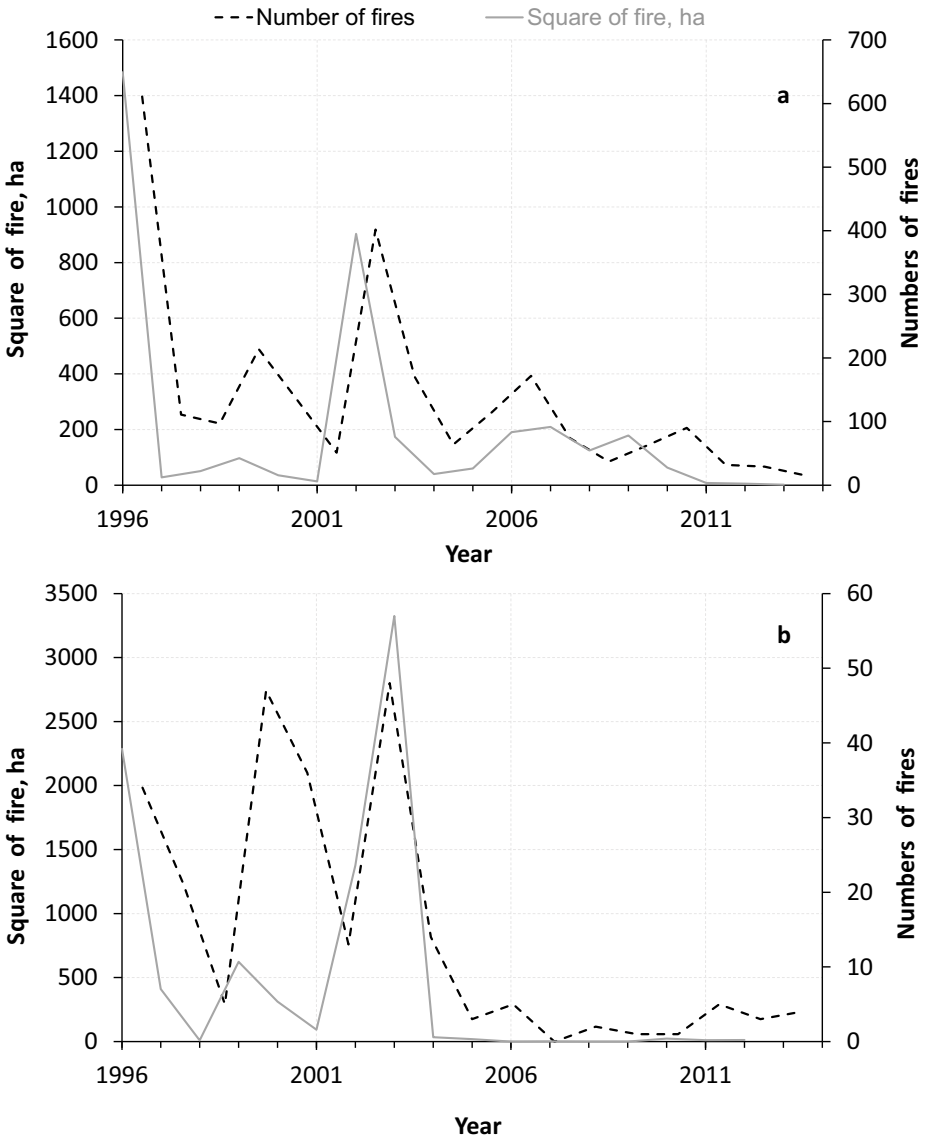

Fig. 1. Long-term dynamics of fires in the areas of radioactive contamination of the Gomel region (a) and in the CEZ (b).

Some studies indicate the increase of volume activity of ^{137}Cs in the summer months (Kulan, 2006). The risk of forest fires is higher in the period from April to September (Zibtsev *et al.*, 2015). In the winter season, forest fires are completely absent. Therefore, any increase in activity of ^{137}Cs in the air in the winter season is not related to forest fire on contaminated territories and depends on other causes. Fire statistics performed by the National monitoring service of Belarus shows that more than 2480 forest's and grassland's fires leading to burning area of more than 3600 ha occurred in Gomel region between 1996 and 2014 (Fig. 1a), including areas with the level of contamination by ^{137}Cs above 555 kBq m^{-2}. Time series of the number of wildfires in CEZ between 1996 and 2012 is shown in Figure 1b. More than 90% of all cases occurred because of anthropogenic factors. In addition, sanitary cutting in the contaminated areas was not conducted for more than 20 years. This led to a significant accumulation of dead organic matter in the restricted access areas (Mousseau *et al.*, 2014).

The objective of this study is to analyze the air pollution by ^{137}Cs as the result of biomass burning emission and transport of radionuclides during forest fires and to estimate a potential threat for human health. We also present an assessment method the transfer of radioactive pollutants with smoke of forest fires on the territories contaminated by radionuclides. The estimation process consists of two stages. The first one based on empirical data obtained from the fire experiment. The second stage based on analysis of climatic, environmental and historical data obtained from continuous datasets in Belarus since 2005.

2 Materials and methods

The research methodology is based on the determination of the volume activity of ^{137}Cs in smoke emissions close to the fire and 10 km away. In order to study the determination of the volume activity of ^{137}Cs in smoke emissions, the fire experiment was applied.

Table 1. Characteristics of the experimental sites.

	Site #1	Site #2	Site #3
^{137}Cs contamination density, kBq m^{-2}	2960	962	560
Background airborne activity, mBq m^{-3}	5.6	2.7	3.1
Dry fuel stock*, kg m^{-2}	3.0	2.6	2.7

* It includes forest litter, dry branches (diameter less than 10 mm), leaves (needles), and dry bark fragments (according to the classification, described in Kurbatskiy, 1962).

2.1 Characterization of objects and sampling

Sampling of fuel materials was carried out at three experimental forest sites located in areas with different levels of radioactive contamination density. The first site is located near the former village Kruki (*N* 51°31′24,6″, *E* 30°19′19,6″), at a distance of approximately 8 km north of the ChNPP. Dose rates were 5.0 ± 0.19 μSv h^{-1} in Kruki (air dosimetric survey measured at 1 m above ground). The second and the third experimental forest sites are located 150 km NE (*N* 52°61,941′, *E* 31°27,708′) of the ChNPP and belongs to the forestry of Vetka. Dose rates were 0.82 ± 0.02 μSv h^{-1}. All experimental sites are largely characterized by dry sandy soils, and Scotch pine (*Pinus sylvestris*) forests. The average age of the trees in the forest was 50 ± 5 years. The general characteristics of the three sites are summarized in Table 1.

To collect the fuel material samples, plots (50 by 50 m) were selected for each forest site. The fuel materials include samples of forest litter with two horizons (A_0L, and A_0F + A_0H). It also includes dry plants, branches with needles (diameter less than 10 mm) and fragments of dry bark.

For the determination of contamination density, 10 soil samples were collected from the each site. Samples were collected in the depth of 20 cm with a 5 cm diameter core sampler. Samples of forest litter were collected with square sampler (50 by 50 cm) at ten points on each plot. Litter was separated into layers: A_0L (light fraction of forest litter), and A_0F + A_0H (mixture of the slightly decomposed forest litter and well humified soil organic matter). A_0F and A_0H layers were collected together. Samples of plants, small branches with a diameter less than 10 mm and dry cones were collected over 1 m^2 area in three replicates. All samples were dried and weighted in the laboratory. The stock of fuel for each experimental site was calculated by summing the dry weight of all samples to 1 m^2.

2.2 Fire experiment

The experiment was carried out in controlled conditions using the smoke chamber. The smoke chamber was designed by employees of laboratory of radioecology specially (Institute of radiobiology of NAS of Belarus) for sampling and analysis of substances in smoke including radionuclides. The smoke chamber is a metallic cylinder with two compartments inside (Fig. 2). The upper compartment is used for the combustion of fuel materials with different levels of radioactive contamination. The lower compartment is used for collecting ash. The chamber is also equipped with an exhaust pipe used to collect aerosols.

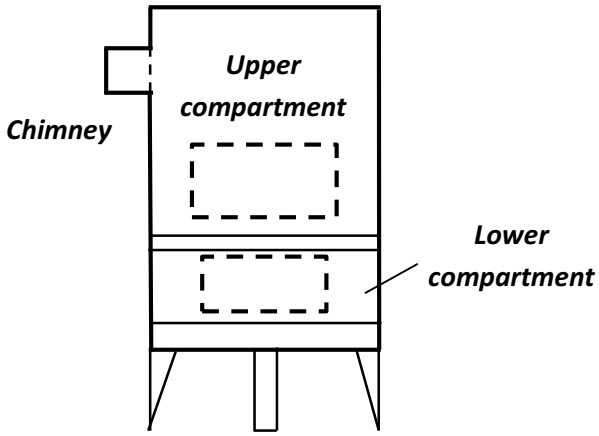

Fig. 2. Scheme of the smoke chamber.

Combustibles materials are incinerated at temperatures above 600 °C. At this temperature, a concentrated stream of smoke is created. For radioactive aerosol sampling, an aspirator with an output of 200 L min^{-1} was used. Duration of sampling was 10 ± 2 min (flow rate 200 L min^{-1}). Uncertainty was equal 5%. Aerosols were sampled on perchlorovinyl fiber filters with a filtration surface of 10 cm^2. After combustion of the material ashes were removed and placed in glass vessels. Fiber filters were placed in a clean polybags. To minimize the possibility of cross-contamination, the chamber was thoroughly cleaned of combustible materials residues and ash.

During the experiment three combustion cycles of materials from the each site was made. The total amount of samples was 9 for ashes and 27 for aerosols. The values of airborne activity concentration A_V of ^{137}Cs is calculated as follows:

$$A_V = \frac{A_f}{V}(\text{Bq m}^{-3}), \quad (1)$$

where A_f is the activity measured in the analytical filter (Bq) and V is the air volume passed through the filter (m^3).

2.3 Sample preparation and activity measurements

Aerosol filters were prepared by serial (three times) dissolving in a solution of 7M HNO_3. After each iteration, the separation of the filtrate was made. After that, the filtrate was adjusted to a volume of the measuring cuvette with 1M HNO_3.

Soil, plant and forest litter samples were measured air-dried. Each sample was placed in cylindrical plastic vessels with complete filling of geometry. Two geometries were used for measurements: one with a diameter of 14.5 cm and 11 cm height and the other with a diameter of 7 cm and 3.2 cm height.

The activity concentration of ^{137}Cs in all samples was measured with the γ-spectrometric complex CANBERRA (USA), which is equipped with the Ge-detector (model GX2018). The energy resolution of the detector is 1.8 keV for the ^{60}Co-line at 1.33 MeV. Detection efficiency of spectrum for energy 1.33 MeV is 22.4%.

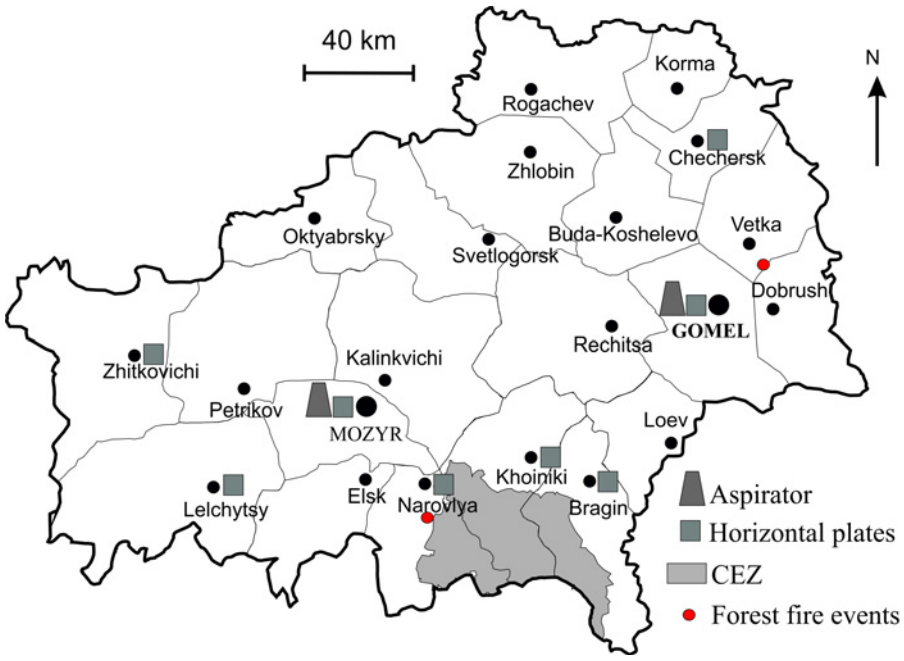

Fig. 3. Monitoring network in Gomel region.

2.4 Determination of atmospheric pollution by ^{137}Cs

Determination of air pollution by ^{137}Cs from forest fires on analysis of radiation monitoring data for the period from 2003 to 2013 was based. The data on the dynamics of forest fires in the zones of radioactive contamination of the Gomel region, and information on meteorological conditions for interpretation were also used. Data were compared in compliance with:

- distance from the fire site to the radiation control station shall not exceed 10 km;
- fire area shall not be less than 0.1 ha;
- fire site must be located on the territory having a level of radioactive contamination by ^{137}Cs over 555 kBq m^{-2};
- wind direction must be the same with the direction from forest fires to the radiation control center.

The atmospheric pollution by radionuclides in Belarus is controlled by center of radioactive contamination control and environmental monitoring. In accordance with the National monitoring network, there are a few stations of radiation control in the Gomel region. Aerosol sampling stations in Gomel and Mozyr are equipped with Petryanov filters and air pump with a flow rate of 1000 m^3 h^{-1}. The stations in other cities have only horizontal plates (Fig. 3).

In comparative database, we generally use the data on the density of radioactive contamination of forest site, area, type and intensity of the fire and the wind direction. The number of entries in the database was 426. The approach that we used is also based on calculation of an activity index defined as the ratio between airborne activity concentration during fire and a reference background concentration. The activity index represents the excess of volume activity of ^{137}Cs in the air on the day of the fire over the reference values of volume activity of ^{137}Cs in the air (in the days without fire). The absence of fire in the winter season allowed us to use data on the radioactive contamination of the air by ^{137}Csin winter season of each year as a reference values.

The activity index values were achieved by using the formula:

$$I_A = \frac{A_{Vi}}{A_0}, \quad (2)$$

where A_{Vi} is the activity concentration of ^{137}Cs in air on the day of the forest fire (Bq m^{-3}) and A_0 is the reference values of ^{137}Cs volume activity in air in the days without fire (Bq m^{-3}).

3 Result and conclusions

The forest litter is the primary main combustion conductors during the forest fires (Kurbatskiy, 1962). It also includes dry grass and mosses, lichens, fallen leaves, branches, bark fragments, etc. Pine phytocenoses have well-developed forest litter (3–6 cm). Characteristics and the measured ^{137}Cs activity in the samples from the experimental forest sites are presented in Table 2.

Data in Table 2 show that more than 90% of ^{137}Cs is associated with the forest litter. The main source of litter is litterfall, which is also a good conductor of burning. The contents of ^{137}Cs in the components of the litterfall vary from 10% to 15% of its total activity in combustible materials. More than 50% of cesium activity in the litterfall are associated with needles.

3.1 Concentration of ^{137}Cs in combustible products during the fire experiment

The data shows that airborne activity of ^{137}Cs increase with increasing of ^{137}Cs activity concentration in combustible materials (Fig. 4). The experimental data are

Table 2. Characteristics of various compartments of forest fuel materials.

	Activity concentration of ^{137}Cs, kBq kg^{-1}	Biomass density*, kg m^{-2}	Contamination density, kBq m^{-2}
Radioactive contamination density of territory 2960 kBq m^{-2}			
Forest litter	154.9 ± 13.2	2.6 ± 0.4	402.7 ± 32.1
Fallen branches	16.9 ± 0.2	0.08 ± 0.01	1.4 ± 0.2
Bark + needles + cones	32.1 ± 1.2	0.24 ± 0.02	7.7 ± 0.8
Total		3.0 ± 0.4	411.8 ± 33.1
Radioactive contamination density of territory 560 kBq m^{-2}			
Forest litter	61.7 ± 5.2	2.5 ± 0.3	154.2 ± 19.1
Fallen branches	1.32 ± 0.08	0.034 ± 0.005	0.044 ± 0.005
Bark + needles + cones	7.2 ± 0.4	0.13 ± 0.02	0.97 ± 0.09
Total		2.7 ± 0.3	155.3 ± 19.2
Radioactive contamination density of territory 962 kBq m^{-2}			
Forest litter	85.5 ± 10.1	2.4 ± 0.1	205.2 ± 25.4
Fallen branches	3.07 ± 0.19	0.044 ± 0.006	0.13 ± 0.02
Bark + needles + cones	10.8 ± 0.6	0.15 ± 0.01	1.62 ± 0.13
Total		2.6 ± 0.1	206.9 ± 25.5

* Dry weight.

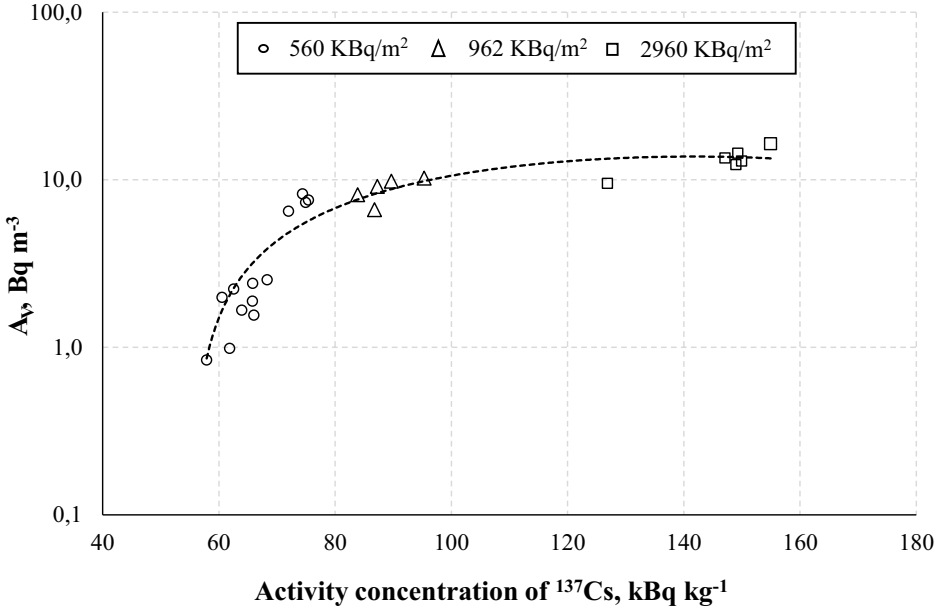

Fig. 4. Airborne activity concentration of ^{137}Cs during the experiment.

fitted with a two-order polynomial ($r^2 = 0.89$). Averaged values of airborne ^{137}Cs concentrations during combustion of fuel materials taken from territory with contamination density of 560 kBq m^{-2} was one order of magnitude lower than those taken from territory having contamination density of 2690 kBq m^{-2}, *i.e.*, 4.8 times more. Activity concentration of radionuclides in aerosols depends on several factors: the contamination density of fuel materials, combustion intensity and duration.

The measured specific activity of ^{137}Cs in ash samples was 2–3 order of magnitude higher than the specific activity of ^{137}Cs in combustible materials (Amiro *et al.*, 1996; Yoschenko *et al.*, 2006).

According to the radiation safety standards the average permissible annual volume activity for workers and for population is 1.7 kBq m^{-3} and 27 Bq m^{-3}, respectively (Radiation Safety Standards in Belarus, 2000). Airborne concentration of ^{137}Cs produced by the combustion of forest fuel materials with contamination density over 400 kBq m^{-2} can exceed the permissible activity levels of ^{137}Cs in air for population (46.7 Bq m^{-3}), but not enhance the permissible activity levels of ^{137}Cs for workers and firefighters. During the combustion of forest materials with radioactive contamination density 154 kBq m^{-2} a significant enhancement of allowable activity levels of ^{137}Cs in air was not detected.

Table 3. Monthly activity of fallout on horizontal plates (stations of radiation control in Gomel), Bq m^{-2}.

Month	Year					
	2006	2007	2008	2009	2010	2011
January	0.65	0.32	0.44	0.65	0.62	0.63
February	0.46	1.18	0.54	0.47	0.59	0.38
March	0.52	0.77	0.64	0.76	0.71	0.53
April	0.76	0.67	0.73	0.58	0.62	0.56
May	0.68	0.90	0.57	0.58	0.7	0.43
June	0.67	0.57	0.30	0.75	0.52	–
July	0.48	0.77	0.49	0.55	0.53	0.46
August	0.84	0.45	0.61	0.36	0.65	0.61
September	0.63	0.43	0.50	0.67	0.65	0.54
October	0.87	0.77	0.72	0.93	0.54	0.59
November	0.67	0.70	0.45	0.80	0.62	0.69
December	0.77	0.61	0.56	0.56	0.57	0.67

3.2 Dynamics of ^{137}Cs airborne concentration during the fire season

Comparative analysis of ^{137}Cs fallout on horizontal plates shows that there is no significant difference between summer and winter seasons. Thus, the data show that the mean monthly activity of fallout on horizontal plates near Gomel in winter and summer during 2008 were equal (0.57 ± 0.09) and (0.53 ± 0.08) Bq m^{-2} per day, respectively ($t_{05} = 1.75$, df = 10). Since 2006, the fluctuations in the density of radioactive contamination fallout in summer and winter are similar (see Tab. 3).

On the other hand, the mean year fluctuations of ^{137}Cs airborne concentration show a significant increase of cesium contents in air from April to September each year. As it shown in Figure 5, the ratio between the volume activity of ^{137}Cs in the air in summer and winter season was 1.26 ± 0.06 for more than 7 years. The single values of monthly volume activity in comparative database exceeded reference values by 1.5 or even

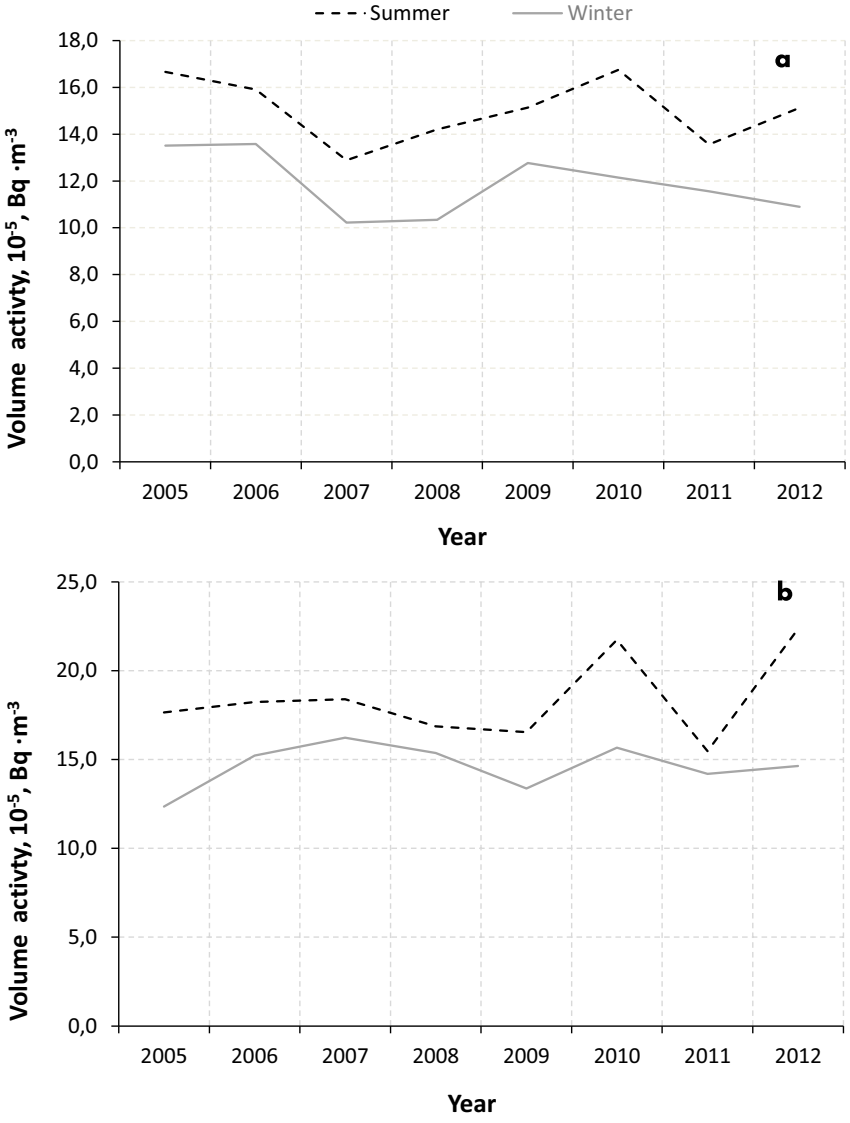

Fig. 5. A year-to-year variation of ^{137}Cs airborne concentration in Gomel (a) and Mozyr (b).

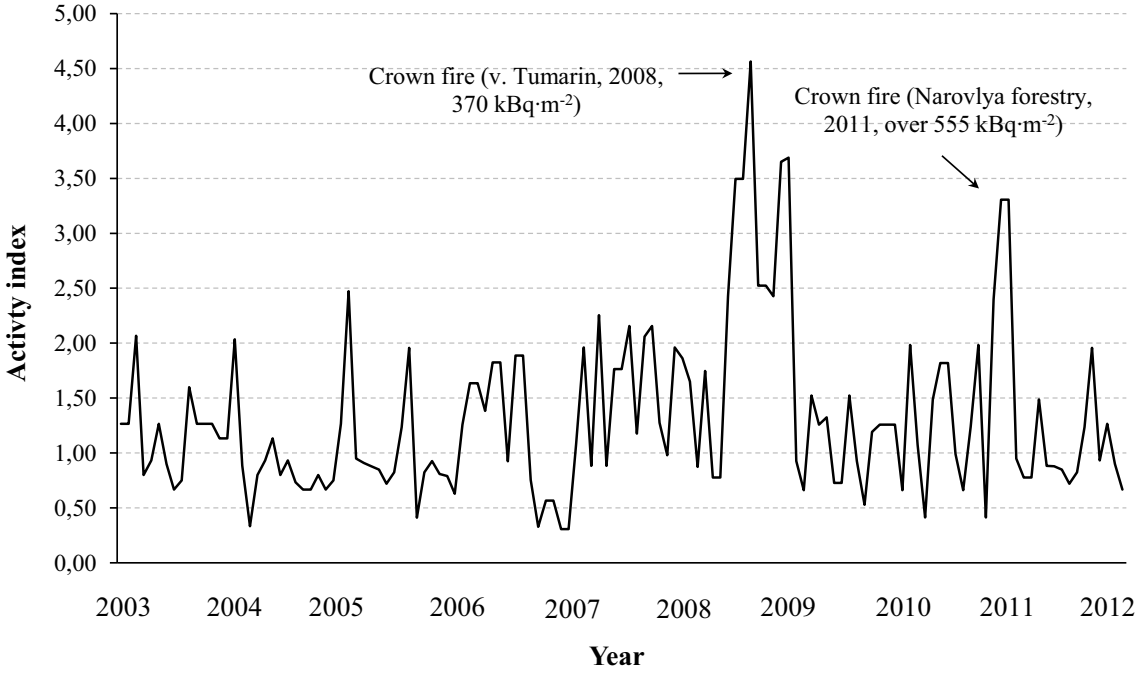

Fig. 6. Time's series of activity index of ^{137}Cs after the forest fires (in the brackets: the location of fire source, year, and contamination density by ^{137}Cs).

2 times. The main reason of such ratio is the full absence of forest fires in the winter.

The only possible source of the air contamination can be associated with wood heaters activity and stove heating usage during the winter season. However, the contribution of this factor in the air radioactive contamination cannot be estimated authentically, because of the absence of significant statistical data. In addition, according to the hygienic standards in Belarus (National Hygienic Standards, 2001), the activity concentration of ^{137}Cs in the firewood cannot exceed the permissible level of 740 Bq kg^{-1}. Usage of any materials with more or less large quantities of ^{137}Cs is strongly forbidden. However, the issue may be of interest for further research.

The activity index indicates significant enhancement of airborne concentration of ^{137}Cs during the large wildfires on the territories with contamination density over 370 kBq m^{-2}. As a result of ground and crown forest fires with high intensity, the background values of ^{137}Cs airborne concentration may be increased by three or four times. Figure 6 shows the time series of the activity index during forest fires in Gomel region.

In all cases with high ^{137}Cs activity index, the distance from the source of forest fire did not exceed 10 km. Thus, during the crown fire near the village of Tumarin (Vetka forestry, 52°30′12.2″*N*, 31°13′34.6″*E*) over 60 ha of pine forest were burned during the first day and 45 ha – in the second day of fire. The total burning area was larger than 100 ha. ^{137}Cs contamination density of territory around the fire was 370 kBq m^{-2}. The wind direction during the fire was the same than the direction from the source of fire to the stations of radiation control (Gomel). The average daily activity of ^{137}Cs in the air on the stations of radiation control was $(47 \pm 5) \times 10^{-5}$ Bq m^{-3}, which exceeds four times the background values (the average monthly values of background activity by ^{137}Cs was $(12 \pm 2) \times 10^{-5}$ Bq m^{-3}). The lowest background value along 2008 counted by using percentile 10 thresholds was equal to 4.0×10^{-5} Bq m^{-3}.

Another case of crown forest fire was registered on territory with contamination density by ^{137}Cs over 555 kBq m^{-2} (Narovlya Forestry, 51°42′5″ *N*, 29°37′35″ *E*). The activity index was 3.5 (the stations of radiation control in Mozyr).

The analysis of a year-to-year fluctuations of activity index show that its value depends on several factors such as radioactive contamination density of territory and forest fuel components (forest litter, small branches, needles and pinecones), forest fire area and its intensity, wind speed and direction.

In this way, experiments showed that the higher the radioactive contamination density of the fuel components, the higher the airborne concentration of radionuclides in the case of forest fire. Radionuclide fallout from the smoke plume on horizontal plates is low and the level of radioactive contamination is not changed practically. However, during the large wildfires on the territories with high values of radioactive contamination density the airborne concentration of radionuclides may be three or four times higher than the background values and exceed the average permissible annual volume activity for population. With a fair wind, the high values of airborne concentration of radionuclides also can be detected at medium range distance (10 km) from the fire source.

The fire source can be located at the distance up to 10 km from the station of radiation control. Additionally, dispersal of the smoke cloud by wind can lead to a decrease of radionuclides concentration in depositions. For these reasons, the horizontal plates sampling method has low accuracy at medium range distance. The data of radioactive fallout density in winter and summer can be similar within the statistical error. To obtain significant values of radioactive fallout density the horizontal plates must be set on short distance (up to 100 m) from the fire source as it indicated in Yoschenko *et al.* (2006).

The methodological approach of estimation of radionuclides release from fire source we presents above allowed us to use the empirical data for annual effective dose calculation for firefighters and for local population at the medium range distance. Prediction of doses for firefighters and amount of radionuclides that potentially can migrate with air masses outside of fire location are the main actions of the project in the framework of Belorussian State Program of scientific research 2016–2018. It is necessary to develop scientific recommendations to improve the efficiency of wildfires prevention at the territories contaminated by radionuclides (especially for territories near large cities with more or less low levels of contamination density). The outcomes data are also of great importance to inform the population and reducing of social and psychological stress in the society caused by the influence of mass media and the lack of scientifically based models of wildfire and radionuclides release.

Acknowledgments. The presented research was carried out within the framework of the project supported by the Belorussian State Program for Basic Research "The potential of natural resources". We gratefully acknowledge the anonymous reviewers for their careful reading, critical comments and helpful suggestions on the paper.

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Cite this article as: Dvornik AA, Klementeva EA, Dvornik AM. 2017. Assessment of ^{137}Cs contamination of combustion products and air pollution during the forest fires in zones of radioactive contamination. *Radioprotection* 52(1): 29–36.